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Invited Article

OVERVIEW ON HYDROGEN RISK RESEARCH AND DEVELOPMENT ACTIVITIES: METHODOLOGY AND OPEN ISSUES

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ABSTRACT

During the course of a severe accident in a light water nuclear reactor, large amounts of hydrogen can be generated and released into the containment during reactor core degradation. Additional burnable gases [hydrogen (H_2) and carbon monoxide (CO)] may be released into the containment in the corium/concrete interaction. This could subsequently raise a combustion hazard. As the Fukushima accidents revealed, hydrogen combustion can cause high pressure spikes that could challenge the reactor buildings and lead to failure of the surrounding buildings. To prevent the gas explosion hazard, most mitigation strategies adopted by European countries are based on the implementation of passive autocatalytic recombiners (PARs). Studies of representative accident sequences indicate that, despite the installation of PARs, it is difficult to prevent at all times and locations, the formation of a combustible mixture that potentially leads to local flame acceleration. Complementary research and development (R&D) projects were recently launched to understand better the phenomena associated with the combustion hazard and to address the issues highlighted after the Fukushima Daiichi events such as explosion hazard in the venting system and the potential flammable mixture migration into spaces beyond the primary containment. The expected results will be used to improve the modeling tools and methodology for hydrogen risk assessment and severe accident management guidelines. The present paper aims to present the methodology adopted by Institut de Radioprotection et de Sûreté Nucléaire to assess hydrogen risk in nuclear power plants, in particular French nuclear power plants, the open issues, and the ongoing R&D programs related to hydrogen distribution, mitigation, and combustion.

1. Introduction

During severe accidents (SAs) in a nuclear power plant, hydrogen can be produced from exothermal oxidation of fuel cladding or fuel assembly canisters, other hot metallic components, and molten core–concrete interaction (MCCI) after failure of the reactor pressure vessel and melt relocation to the reactor pit if in-vessel retention measures failed. A large

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amount of carbon monoxide may also be produced during MCCI in addition to hydrogen and other gases. The hydrogen released into the containment via a reactor cooling system (RCS) break or through the pressurizer safety valves or during corium-concrete interaction is transported by convection loops arising essentially from the released hot steam/gas or initiated by condensation of steam on cold walls. Depending on the level of mixing in the containment atmosphere, the distribution of hydrogen can be homogeneous or stratified. If considerable hydrogen stratification exists, local concentration of hydrogen and carbon monoxide may become a safety concern because pockets of high hydrogen and carbon monoxide concentrations may lead to combustion, flame acceleration (FA), and eventually deflagration to detonation transition (DDT) if the combustible mixture is ignited. Furthermore, the hydrogen distribution may be affected by safety systems such as spray or coolers that are widely used in many reactors to reduce the containment pressure and to provide heat removal by steam condensation on water droplets or cold surfaces. These measures may homogenize the hydrogen distribution in the containment because of enhanced mixing, but they can also significantly reduce the steam concentration, which may lead to more combustible gas mixture compositions.

Several methods [1] can be proposed to reduce the hydrogen concentration in the containment: (1) deliberate ignition, (2) consumption or recombination of hydrogen by passive autocatalytic recombiners (PARs), (3) combining ignition and recombination measures, (4) replacing oxygen (air) with an inert gas [typically nitrogen (N_2)] during normal plant operation, (5) diluting the atmosphere by having, by design, a large containment volume or by injecting an inert gas, or (6) releasing hydrogen by containment venting. Hydrogen risk management can be implemented by one or a combination of the previous measures.

The choice of a mitigation strategy depends primarily on the design of the containment. For pressurized water reactors with a large dry containment, the strategy usually consists of combining a large free volume to allow dilution, a high value of the design pressure, and the use of mitigation means such as PARs to consume hydrogen. This strategy is adopted in all French pressurized water reactors. The implementation of PARs were performed following rules and criteria that allow (1) the reduction of the possibility of hydrogen accumulating to flammable concentrations, (2) the minimization of the flammable gaseous volume, and (3) the prevention of an increase in the hydrogen levels from flammable to detonable mixture concentrations. For French reactors, the adopted criteria consist of keeping the hydrogen concentration mean value below 8%vol to avoid complete combustion and the local hydrogen concentration value under 10%vol to avoid FA. These criteria were established to achieve the safety objective set for French reactors, which is to practically eliminate situations that could yield an early containment failure with large early releases. Hydrogen combustions that can challenge the containment integrity consequently have to be avoided. Therefore, for an auxiliary building, hydrogen combustion should not challenge any safety function and system.

To evaluate the efficiency of such mitigation strategies, the methodology usually adopted is based on the prediction of hydrogen distribution inside the containment while taking into account the mitigation and safety systems effect and the prediction of pressure and temperature loads that are induced by the combustion. To cover all conceivable situations of a severe accident, including in- and ex-vessel phases, research and development (R&D) efforts are still needed to improve the methodology described in more detail in the next paragraph.

2. Hydrogen risk assessment methodology

To fulfill the requirements for the management of the hydrogen risk in the nuclear power plant (NPP) mitigation measures have been implemented in various NPPs containments to remove hydrogen or to control the hydrogen concentration inside the containment. The appropriate mitigation measures is then designed, based on the results of numerical simulations and dedicated experiments. The methodology usually adopted to assess the hydrogen risk in the reactor building and in the auxiliary buildings consists of the following seven steps:

Step 1: Modeling of the plant design.

The starting point of any analysis is the selection of the plant and geometrical modeling of the containment or the auxiliary building. This step aims to describe adequately the building geometry and volumes (including compartments) and the safety system considered (e.g., PARs, spray, coolers), which influence hydrogen distribution inside the reactor containment or auxiliary building.

Step 2: Selection of relevant accident scenarios.

The representative or bounding in terms of hydrogen-produced (mass and kinetics) severe accident scenarios for hydrogen assessment are generally identified from SA code calculations such as ACTEC calculations, Probabilistic Safety Analysis (PSA) level 1 and 2 or other analyses. The evaluation of the associated hydrogen production rates and release into the reactor building is usually derived from parametric code calculations and best estimates calculations considering remaining uncertainties on hydrogen production processes.

Step 3: Evaluation of the containment atmosphere condition.

During the accident transient, the temperature, pressure, and gas composition in the different regions and volumes of the containments are determined, and account for the presence of the mitigation systems. For this purpose, both lump parameter (LP) and computational fluid dynamics (CFD) codes are often used to simulate the transient. The CFD codes are usually used for deeper analysis in short-term temporal windows (e.g., hydrogen release phase, scenarios with local accumulation, and expected stratification).

Step 4: Evaluation of the time evolution of a flammable hydrogen-air-steam cloud.

The flammability of the containment gas mixture depends on its temperature, pressure, and composition as shown in Fig. 1.

In practice, the point representing the mixture composition (hydrogen, air, steam) on the Shapiro diagram (Fig. 2) is generally used to determine whether the mixture is flammable. In this diagram, the zones of ignition (yellow) and detonation (orange) are delimited by the exterior and interior curves, respectively. The detonation limit is not an intrinsic property; it is geometry-dependent. The red lines are simplified combustion limits that are often used in codes.

Step 5: Evaluation of the propensity of a premixed flame to propagate inside a building.

Under the effect of hydrodynamic instabilities and turbulence (caused primarily by obstacles in the flame's path), an initially laminar deflagration with a flame velocity of approximately 1 m/s may accelerate. Fast combustion regimes may also develop, and involve rapid deflagration (a few hundred m/s), DDT, and detonation (greater than 1000 m/s). These combustion regimes may generate high pressure loads that could endanger the integrity of the containment or the safety system's integrity. For this purpose, FA criteria (called the "σ" criteria) are used. The symbol σ stands for the mixture's expansion factor, which is the ratio of unburnt gas versus burnt gas densities at constant pressure. It is an intrinsic property of the mixture. The critical value σ^*, beyond which FA is possible, depends on the initial gas composition and temperature and flame stability.

Step 6: Evaluation of the pressure and thermal loads generated by combustion.

Two configurations are distinguished:

1. If the FA criteria are not met, dynamic pressure loads are excluded and the pressure load is evaluated by considering the adiabatic isochoric complete combustion (AICC) process.
2. If the FA criteria are met, the induced combustion loads is evaluated using the most appropriate combustion models, primarily using modeling implemented in CFD codes.

Step 7: Evaluation of the structure response to pressure and thermal loads generated by combustion.

The structural resilience of the containment can be evaluated using dedicated dynamic structural response codes or based on criteria used to judge whether the containment integrity is maintained.

As mentioned previously, the evaluation of the hydrogen risk is based on the use of validated tools for hydrogen distribution, hydrogen mitigation, and combustion and for the assessment of the structure response to pressure and temperature loads which result from hydrogen combustion. In the past, comprehensive R&D programs were launched to assess and improve the predictability of such tools. In the following paragraph, a short overview on the outcomes of these programs is discussed.

3. Overview of recent international R&D activities to improve hydrogen risk assessment

In the past few decades, extensive studies have been performed and are still continuing in the international nuclear community to assess the hydrogen risk in SAs. A large number of projects sponsored by the Organisation for Economic Cooperation and Development (OECD) or the European Commission have been developed to address the remaining research items on hydrogen and many state-of-the-art reports have been published on various aspects of hydrogen issues (e.g., generation, distribution, combustion and mitigation). A detailed overview on the project thus performed is described in the OECD status report on Hydrogen Risk Management and Codes published recently [25]. A brief overview of the recent R&D activities related to hydrogen distribution, hydrogen mitigation, and hydrogen combustion is provided.

3.1. *Hydrogen distribution*

In 1999, the OECD *State Of the Art Report on Containment Thermal Hydraulics and Hydrogen Distribution* was published [2] and gave an overview on the code capabilities to predict pressure, temperature, and gas concentration-distribution inside a containment under severe accident conditions. The report

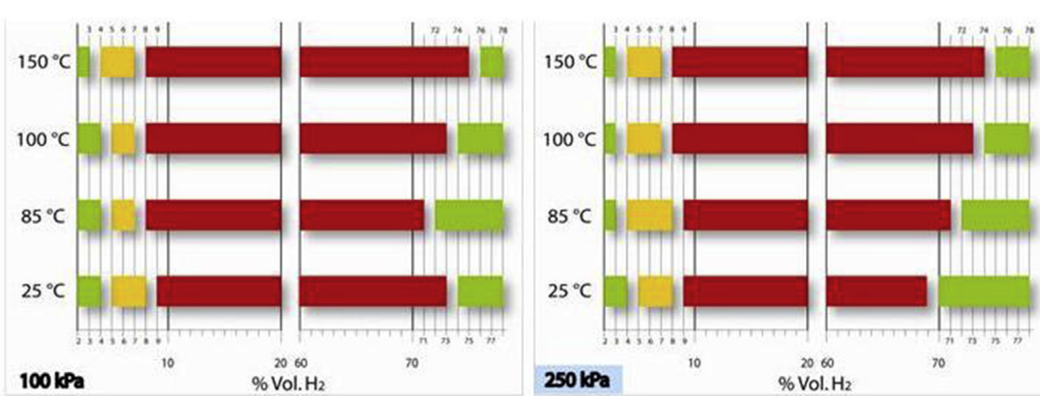

Fig. 1 – Flammability limits diagrams of hydrogen (H_2)/air mixtures between 25°C and 150°C at an initial pressure of 100 kPa and 250 kPa. Green indicates no flame propagation; orange, upward propagation; and red, full propagation.

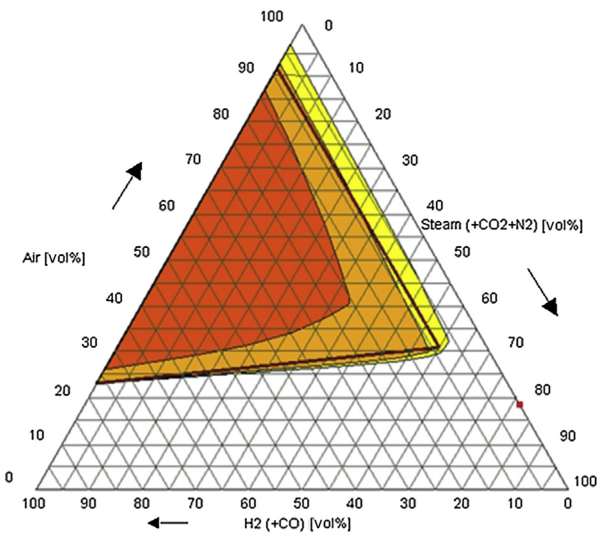

Fig. 2 – Shapiro diagram for hydrogen–air–steam mixtures at 1 bar and ambient. CO_2, carbon dioxide; CO, carbon monoxide; N_2, nitrogen.

also described the experimental activities performed in the 1980s and 1990s that investigated phenomena related to containment thermal hydraulics and hydrogen distribution. Furthermore, this report discussed code development and application activities, particularly ISP-23, ISP-29, ISP-35, and ISP-39 benchmarks against the Heiss-Dampf Reaktor (HDR) and Nuclear Power Engineering Corporation (NUPEC) hydrogen mixing and distribution tests. The conclusions of this report highlighted the remaining uncertainties from experimental and models development and highlighted the need for refined measures to validate lumped parameter and CFD codes.

To resolve the most important remaining issues for SAs [3], the European Commission (EC) established a 4-year Severe Accident Research NETwork of Excellence (SARNET) project in 2004. The containment behavior was one of the topics addressed. The influence of containment sprays on atmosphere behavior was investigated through benchmark exercises based on experiments performed in TOSQAN (IRSN, France) and MISTRA (CEA, France) facilities [4]. It was concluded that the level of validation obtained was encouraging for the use of spray modeling for risk analysis. However, some more detailed investigations were recommended to improve model parameters and decrease the uncertainty for containment applications.

The *International Standard Problem ISP-47 on Containment Thermal-hydraulics* [5] was later launched with the main objective of assessing the capabilities of the LP and CFD codes in the area of containment thermal hydraulics. Data generated in three experimental facilities TOSQAN, MISTRA and Thermal-hydraulic, Hydrogen, Aerosol and Iodine (THAI) (Becker Technology, Eschborn, Germany) were used for benchmarking codes in steady-state and transient conditions. In addition to the pressure transients and gas temperature fields studies in the former benchmark exercises, detailed gas velocity and gas concentration (air, steam and helium) fields were measured and analyzed for the first time in such an exercise. It was concluded that further work was needed in code development, in the establishment of nasalization rules, and in code user training to achieve more accurate predictive capabilities for containment thermal hydraulics and atmospheric gas/steam distribution. Uncertainties in the modeling of condensation, gas stratification, and jet injection appeared to be the major issues.

Following the ISP47 conclusions, the OECD-THAI project was initiated with the aim of answering the open question regarding the substitution of hydrogen with helium in experimental tests and to assess the ability of LP and CFD codes to predict the hydrogen stratification build up and break-up. The investigation of the break-up of hydrogen stratification in an light water reactor containment caused by heat and mass sources and sinks, and by the effect of the activation of a safety system (e.g., spray, heat source simulating a recombiner, containment cooler) was also addressed in the framework of the OECD/SETH2 Project. The experimental investigations were performed in the multi-compartment, large-scale PANDA (PSI, Villigen, Switzerland) and Mitigation and Stratification (MISTRA) facilities, which are equipped with CFD-grade instrumentation. The effect on the hydrogen concentration of the sudden opening of hatches separating a two-compartment containment was also investigated. The accompanying analytical activities performed with various computational tools during the project have already proven the relevance of the experimental database for the assessment of advanced LP and CFD code capabilities.

More recently, the European Union and Russian State Atomic Energy Corporation cofunded the 4-year ERCOSAM-SAMARA [6] projects to investigate the hydrogen concentration build up and stratification under stylized postulated accident scenarios, and to investigate the effectiveness of accident management systems (i.e., cooler, spray, and PAR) in destabilizing a stratified hydrogen layer. The experimental investigations were performed in the PANDA, MISTRA, TOSQAN, and SPOT (JSC, Russian) facilities. The use of different facilities allows insight on scaling effects and to draw general conclusions that are not specific to a particular containment design. In parallel to the experimental program planning, pre- and post-test calculations are being performed by the member countries to assess the experimental conditions, evaluate the test results, and verify codes/models currently used in reactor safety analysis.

The aforementioned projects demonstrate the ability of multicompartment and multidimensional computational codes to predict gas mixing inside the reactor containment, which allows their use for reactor application as in reports by Bleyer et al. [7] and Caroli et al. [8]. However, some remaining issues related to safety systems (spray, PARs, venting) effect on gas mixture still need additional investigation. These issues were highlighted also in the French Safety Authority stress report following Fukushima accident [9].

3.2. *Hydrogen mitigation*

The IAEA report on *Mitigation of Hydrogen Hazards in Water Cooled Power Plants* [10], published in 2001 summarized the concepts for hydrogen mitigation in containments. The conclusions of this report indicated that the choice of measures to mitigate the hydrogen threat depend strongly on the containment design. The implementation of PARs was adopted in the western European countries with different requirements and were based on several experimental results described in a state of the art report on hydrogen recombination [11].

More recently and in the framework of the OECD-THAI and OECD-THAI2 projects [12,15], significant progress in knowledge has been achieved on the behavior (e.g., onset of recombination, recombination rate, and ignition potential) of different PAR designs under severe accident typical conditions. Oxygen starvation significantly reduces the recombination rate. A highly important result is that PAR ignition potential is limited to a relatively small area of mixture compositions in the air-hydrogen steam ternary diagram that are capable of providing the high catalyst temperatures required for ignition. Furthermore, detailed models were recently developed at the IRSN and implemented in SPARK code [13]. The use of this model for the analysis of REKO [14] and THAI [15] experiments allows the determination of the ignition limits induced by PARs. The ongoing collaboration between IRSN and JULICH are permitting the improvement of the SPARK model [13], the investigation of the effect of carbon monoxide on hydrogen recombination [16] and the effect of fire products on recombination start up.

Nevertheless, remaining issues regarding PARs behavior in challenging conditions still need further investigation. These challenging conditions concern PARs efficiency in gas mixture, composed of hydrogen, carbon monoxide from MCCI, steam and low oxygen, and correspond to the late phase of a severe accident. Furthermore, the overall flow conditions inside the containment in the course of a severe accident may be quite different from a quiescent atmosphere. In general, the corresponding flow patterns will be strongly influenced by the location of the release and by components and installations. Especially close to the containment walls, a downward-directed flow is expected to occur in the early accident phase. Because many PARs installed inside a light water reactor are mounted at the containment walls, these PARs will have to establish the internal upward chimney flow against the downward local containment flow. Another relevant issue never investigated before in detail is the influence of the PAR location on the efficiency of the flammable gases conversion.

3.3. *Hydrogen combustion*

The OECD *State of the Art Report on Flame Acceleration and Deflagration to Detonation Transition* [17] addressed the key issue of hydrogen combustion that may occur as a result of SA in NPPs—namely, to predict the type of combustion (e.g., a slow flame, a fast turbulent flame, or a detonation). Despite significant advances in understanding FA and DDT, which had been obtained between 1990 and 1999, the knowledge of FA and DDT is largely empirical and numerical simulation is unable to provide a truly predictive capability for DDT. Many situations arise in which the possibility of FA and DDT cannot be ruled out, but there are no means to predict whether these will actually occur.

Based on these conclusions, the national ENACCEF [18], the European (HYCOM [9] and SARNET [19]) and the OECD projects (ISP49 [20], THAI [12] and THAI2 [21]) were launched with the aim to enhance the knowledge of the flame propagation mechanisms. The results of these projects contribute to enhance the level of knowledge on flammability limits [22], on the FA criteria in semi-confined configuration [19] and in nonhomogenous mixtures [22]. The effect of spray on flame propagation has also been partially addressed [23,24].

The lessons learned from these projects show that the computer codes are able to reproduce the pressure generated by combustion. On the other hand, they do not well predict the flame propagation regimes as observed in the experiments. These are the situations in which the turbulence generated by the interaction between the flame and obstacles is sufficiently high to bring about a transition towards detonation, as was the case for the explosion that occurred in Unit 3 of the Fukushima NPP. These exercises also highlighted the following:

1. The need for turbulence intensity measurements in fresh gases. These data are necessary for turbulent combustion models validation. To date, this information has been missing in most known experiments.
2. The need to study the scaling effect from experimental facilities towards reactor application. The differences between the experimental installations and the reactor containment dimensions (for example a factor of 50,000 between ENACCEF facility of ICARE and the containment of the 900 MWe reactor) do not allow the extrapolation of the lessons learned from the experiments towards the reactor containment. In fact, the scale effect can at the same time accentuate instabilities and decrease the thermal losses (i.e. the losses of the energy of the flame, thus its deceleration, and even for its smothering on smaller scale) and thus promote the acceleration of the flame.
3. In addition to the previous statement, issues regarding flame propagation still need further investigation. These remaining issues concern specifically the determination of flammability limits and FA criteria in MCCI representative conditions (e.g., low oxygen concentration, steam, hydrogen, carbon dioxide, carbon monoxide, nitrogen) and the effect of safety systems (e.g., spray, venting) operation on flame propagation.

4. Ongoing and foreseen R&D activities

In spite of the accomplished efforts of national, European, and international projects to apprehend the whole of the physical phenomena related to hydrogen risk, gaps of knowledge have been indicated [1]. These gaps deal with distribution and combustion of hydrogen, and the dynamic response of containment structures to pressure loads because of hydrogen combustion. This statement had been highlighted recently by the French Nuclear Safety Authority in a report [9] regarding complementary safety assessments after the Fukushima accidents. This report also underlines the need to evaluate the risk of hydrogen deflagration in the fuel building to determine practical and organizational guidelines and the need of appropriate instrumentation to enhance the accident management.

4.1. MYTHYGENE project

Based on these observations, a work program was proposed in the MITHYGENE project to improve the hydrogen risk assessment models and safety management procedures. This program launched in 2013 aims to address the gaps of knowledge concerning the distribution and combustion of hydrogen and its effect on the containment structures. New measurement techniques of hydrogen concentrations in the reactor containment will be developed within the project. Thus, the knowledge gained will be used to improve the accident management guidelines (SAMG) and the predictability of the computer codes used for safety assessment.

The MITHYGENE project is funded by the French Research agency within a framework set by the French national program, Investissements d'Avenir, and supported by industry partners. In addition to the IRSN, the project coordinator, institutional partners, industry, and academia are involved in this project. Institution and university are CEA and ICARE, and the German institute is JÜLICH. The industrial partners are ELTA, AREVA, EDF, and AIR-LIQUIDE.

The MITYHGENE Project offers the unique opportunity to study in a concerted way all the topics dealing with the study of the hydrogen risk, to fill the existing gaps of knowledge and to improve safety strategies in nuclear and industrial installations.

The MITHYGENE project aims at improving the predictability of the numerical tools used for hydrogen risk evaluation, and also at designing and qualifying a prototype of *in situ*, real time and multipoint instrumentation of gas measurement. The results of these actions will be used to interpret the events of Fukushima and to propose an improvement of the mitigation devices and procedures of severe accident management.

For this purpose, the MITHYGENE project includes fundamental and applied topics:

1. Fundamental topics aim at improving models for hydrogen related phenomena and at developing *in situ* and severe accident compatible diagnostics for gas concentrations measurement. Four research topics are then addressed:
   i. The purpose of the first topic is to perform experimental and numerical studies of hydrogen distribution by taking into account the effect of the mitigation means. The results of these studies will improve the capacity of the computer codes to predict situations not yet addressed by national and international programs.
   ii. The second topic aims to perform experimental and numerical studies of hydrogen flame propagation into gaseous atmosphere containing water vapor and water droplets at different initial conditions. The experiments thus performed will fill the lack of data on hydrogen flame propagation in a wet medium. The synthesis of the experimental data will be used to enhance the FA criteria, to improve the models of combustion implemented in the computer codes, and to extend their validation domain.
   iii. The third topic aims to perform experimental and numerical studies of the concrete and metallic structures response to hydrogen explosion loads. Various loads corresponding to various modes of combustion will then be studied. The results will be used to improve the knowledge on dynamic effects generated by combustion on concrete and metallic structures.
   iv. The fourth topic aims to develop a prototypical device for *in situ* and real time measurement of gas composition inside the reactor containment in a SA.
2. The applied topics objective is the enhancement of safety measures related to hydrogen risk based on the knowledge gained from fundamental topics. Three tasks will be performed:
   i. Make industrial the prototype developed in the framework of WP 4.
   ii. Improve the practices and doctrines adopted by manufacturers to prevent hydrogen explosion risk in their own installations.
   iii. Provide a complete explanation of the hydrogen explosion that occurred in the Fukushima Daiichi NPP.

4.2. THAI3 Proposal

The THAI 3 Proposal aims to complement the objectives addressed in framework of MITHYGENE project. Hence, flame propagation in complex multi-compartment geometry will be investigated. In addition, PARs operation under reverse flow will be studied to provide complementary data to the four results of REKO.

4.3. HYCOSAM Proposal

The HYCOSAM Proposal aims to investigate the MCCI-related release of H_2/CO and its distribution: H_2/CO mitigation in the rest of the containment in which already several hundred kilograms of hydrogen, if not consumed otherwise, will be available, and H_2/CO combustion in representative SA conditions. Hence, the effect of venting on gas distribution, the PAR operation under representative MCCI conditions, and flame propagation in related atmosphere will be addressed.

5. Conclusion

The aim of this paper is to present the methodology for hydrogen risk assessment and the related R&D programs. Thus, an overview of the recent national, OECD and European R&D programs dealing with hydrogen distribution, hydrogen mitigation and hydrogen combustion is discussed. These projects aim to enhance the level of confidence of the safety tools used at IRSN by improving their validity domains which allows their use in safety analysis.

Despite the effort, uncertainties and lack of knowledge on PAR behavior and flame propagation mechanism still exist. For this purpose, the national MITHYGENE project was launched and proposals were made (i.e., THAI3 in the framework of OECD and HYCOSAM in the framework of the H2020-European call).

Conflicts of interest

None declared.

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